Interactive comment on
"Attribution of Chemistry-Climate Model Initiative (CCMI) ozone radiative flux bias from satellites" by Le Kuai et al.

We would like to thank the Editorial Review Board and the reviewers for their helpful comments. These feedbacks help us to improve the clarity and readability of the manuscript. Please find our replies following the comments.

Anonymous Referee #2

Review of Kuai et al, "Attribution of Chemistry-Climate Model Initiative (CCMI) ozone radiative flux bias from satellites"

In this paper, Kuai et al. quantify and attempt to interpret model biases in outgoing longwave radiation (OLR) in the wavelength band of strong absorption by tropospheric ozone, 9.6 microns. Using assimilated data, they first construct instantaneous radiative kernels (IRKs) to quantify the sensitivity of OLR at 9.6 μ to four driving variables: tropospheric ozone, water vapor, atmospheric temperature, and surface temperature. The IRKs are calculated at a range of altitudes and across the globe. They then apply the observation-based IRKs to the biases in the four variables in an ensemble of climate models; this step yields the contributions of these variables to biases in OLR at 9.6 μ in each models. The subsequent analysis shows the importance of accurate representations of tropospheric ozone and water vapor in calculating OLR in this wavelength region.

The topic of the paper is important. Understanding the contribution of tropospheric ozone to current and future climate change requires models that can accurately simulate OLR in the relevant wavelengths. The calculations in the paper are fairly straightforward and yield some interesting results – e.g., that the overestimate of tropical tropospheric water vapor present in many models leads to an underestimate of OLR in the ozone wavelength band. But the paper feels like an early draft: much of the analysis in the paper is shallow and the paper is not well written.

I recommend acceptance only after major revisions.
Major comments.
1. A key reason to improve OLR at 9.6 μ is to improve estimates of the radiative forcing from the change in tropospheric ozone from the preindustrial era to the present-day or from the present-day to the future. The manuscript leaves the reader wondering how important are the biases uncovered in the manuscript. Do the biases have much of an effect on ozone forcing estimates over time?

We agree these are all very good points. Yes, the biases we studies have effect on the ozone forcing. We have showed in our early study (Kuai et al., 2017) that the tropospheric ozone radiative effect will be weaker under optical thick sky, such as with cold and wet condition through the secondary effect on ozone IRK. Therefore, the incorrect water vapor or temperature will cause the response in IRK in 9.6 μm band and also effect the other band in OLR. And also the wrong water vapor and temperature will also cause incorrect cloud cover and then ozone loss. Too much or too less ozone and water vapor will have wrong heating rate and effect temperature and cloud. All these will effect the ozone forcing estimates from present day to the past.

In meanwhile don't have the O_3 RF estimation over time from CCMI models to do the similar analysis as Bowman et al., 2013 for ACCMIP. We have raised our request in the most recent CCMI workshop for the RF calculation from the CCMI models. Hope in near future, the data will be available. Then we will do more analysis to understand the uncovered biases and their effect on the ozone forcing over time.

We mentioned in the introduction how 9.6 m band bias would impact the ozone RF over time at Page 3, line 44.

'Aghedo, et al. (2011) applied the TES IRKs to evaluate the O3 radiative effect of chemistry-climate models' O3 biases in the Atmospheric Chemistry Climate Model Inter-comparison project (ACCMIP) (Lamarque, et al., 2013). Bowman et al. (2013) found model OLR bias due to O3 is correlated with RF in the ACCMIP models. This correlation helped to reduce the inter-model divergence in RF by about 30% (Myhre, et al., 2013).'

2. In discussing the sources of biases in the four driving variables, the manuscript provides some detail about how modelers have struggled to improve estimates of tropospheric ozone (e.g., Oman et al., 2011 and 2013; Stenke et al., 2013; Revell et al., 2015). But nothing is said about model biases in the other three variables. Are these biases well-known or new to the community? If known, what are the potential reasons for these biases?

The different biases in the models are known since there are regularly evaluation versus all kinds of observations but the reason are poorly understood. However, the biases in the four driving variables for CCMI study and their radiative biases are new to CCMI community and the reason are not fully understood. To my knowledge, for the CCMI comparison, there are not yet specific analysis about the lower tropospheric water vapor biases and temperature biases. Most of such studies are about past inter-comparison project, like CMIP6 or ACCMIP. For example, Lamarque et al., 2013 did a fairly rough analysis in water vapor biases for ACCMIP (Fig. S4). There is some information on the bias for the GEOSCCM in the technical report by Molod et al., 2012 (see figures 18 and 19). However, this is not the CCMI run specifically. Monks et al., 2015 showed the correlation between the intermodal differences in OH in the POLMIP study with inter-model differences in water vapor. They found GEOSCCM simulated specific humidity has high bias relative to MERRA reanalysis in most troposphere and also in mid troposphere compared to Atmospheric Infrared sounder (AIRS) data. Strode et al., 2015 section 3.4.3 discussed some water vapor biases impact on OH, methane and mehyl choloroform lifetime in GESCCM. Again these analyses are not for CCMI study and didn't provide any potential reasons for the biases.

We add some words at Page 15 line 38 as below.

‘We compute the model biases against reanalysis data for four key variables: O3, H2O, Ta, and Ts. Especially for O3 biases, the newly developed TCR-1 O3 assimilation data (Miyazaki et al., 2015; Miyazaki and Bowman 2017) are, for the first time, used as the state-of-the-art benchmark for tropospheric O3 in models. These specific bias comparisons for the CCMI study cause the modelers to investigate the reasons for these biases and motivate them to improve their simulations. For example, MRI-ESM1r1 shows the reduced LNOx emission help to improve their tropical upper tropospheric O3 and its radiative bias.’

References mentioned above:

Lamarque, J.-F., Shindell, D. T., Josse, B., Young, P. J., Cionni, I., Eyring, V., Bergmann, D., Cameron-Smith, P., Collins, W. J., Doherty, R., Dalsoren, S., Faluvegi, G., Folberth, G., Ghan, S. J., Horowitz, L. W., Lee, Y. H., MacKenzie, I. A., Nagashima, T., Naik, V., Plummer, D., Righi, M., Rumbold, S. T., Schulz, M., Skeie, R. B., Stevenson, D. S., Strode, S., Sudo, K., Szopa, S., Voulgarakis, A., and Zeng, G.: The Atmospheric Chemistry and Climate Model Intercomparison Project (ACCMIP): overview and description of models, simulations and climate diagnostics, Geosci. Model Dev., 6, 179–206, https://doi.org/10.5194/gmd-6-179-2013, 2013.

Molod, A., Takacs, L., Suarez, M., Bacmeister, J., Song, I.-S., and Eichmann, A.: The GEOS-5 atmospheric general circulation model: Mean climate and development from MERRA to Fortuna, NASA, Goddard Space Flight Center, Greenbelt, MD, 2012.

Monks, S. A., Arnold, S. R., Emmons, L. K., Law, K. S., Turquety, S., Duncan, B. N., Flemming, J., Huijnen, V., Tilmes, S., Langner, J., Mao, J., Long, Y., Thomas, J. L., Steenrod, S. D., Raut, J. C., Wilson, C., Chipperfield, M. P., Diskin, G. S., Weinheimer, A., Schlager, H., and Ancellet, G.: Multi-model study of chemical and physical controls on transport of anthropogenic and biomass burning pollution to the Arctic, Atmos. Chem. Phys., 15, 3575–3603, doi:10.5194/acp-15-3575-2015, 2015.

Strode, S. A., Duncan, B. N., Yegorova, E. A., Kouatchou, J., Ziemke, J. R., and Douglass, A. R.: Implications of carbon monoxide bias for methane lifetime and atmospheric composition in chemistry climate models, Atmos. Chem. Phys., 15, 11789–11805, https://doi.org/10.5194/acp-15-11789-2015, 2015.

3. The paper focuses on OLR under clear-sky conditions, which is fine. But cloud cover varies significantly among models. Could variation in clouds also affect model estimates of ozone radiative flux – i.e., OLR at 9.6 μ? The text should include a discussion of how the focus on clear sky conditions affects the conclusions.

We added some discussion based on reviewer’s feedback at Page 3, Line 26:

‘In addition, the presence of clouds is the primary control on atmospheric opacity. Under the cloudy sky conditions, the roles of these variables other than cloud on TOA flux are much weaker. In addition, the variation in clouds could affect model estimates not only of the ozone but also of the flux sensitivity to ozone and other variables. Both ozone and sensitivity will impact the ozone radiative flux but in opposite directions. With cloud cover, the O3 loss will be reduced. That means too much cloud would lead to more ozone production. The presence of the cloud would also cause weaker flux sensitivity to O3 and other variables (IRKs). Therefore, the cloud effect is a battle between the impact on ozone estimation and the radiative sensitivity to ozone (IRK). The differences in cloud variations between the models will complicate the radiative effect. Furthermore, the study of the cloud effect is also currently limited by the global observations of total cloud cover and IRK product under realistic cloud conditions. Without knowing which models have better cloud cover, we benefit from using IRK based on the observed cloud free data by TES. Therefore, here we first try to access the role of O3, H2O, Ta and Ts in the variation of the TOA flux without cloud effect.'

4. The paper is poorly written with many lapses in English, about 1-3 per paragraph throughout. Those co-authors for whom English is a first language should read the manuscript much more closely and rewrite as needed. As is, the manuscript gives the impression that the co-authors did not read it carefully.

We have got the professional grammar check after we addressed all comments from two reviewers.

2

Minor comments.

Page 2. Line 10. Abstract should make clear that the overestimate of atmospheric opacity in the models is due to too much tropospheric ozone and/or water vapor. Also, the large number of significant digits looks suspect.

We addressed reviewer's suggestions by adding words for the reason of the opacity in Page 2, line 8 as below and change the number form 132.9 to 133.

"Overall, the multi-model ensemble mean bias is –133 ± 98 mWm-2, indicating that they are too atmospherically opaque due to trapping too much radiation in the atmosphere by overestimated tropical tropospheric O3 and H2O. Having too much O3 and H2O in the troposphere would have different impacts on the sensitivity of TOA flux to O3 and these competing effects add more uncertainties on the ozone radiative forcing."

Page 2. Line 22. The text should make clear whether these estimates of radiative forcing are instantaneous or adjusted.

This is adjusted radiative forcing. Page 2, Line 21 has been updated as

'Tropospheric O3 adjusted RF ranges widely from +0.2 to +0.6 Wm-2 computed from chemistry-climate model ensembles (IPCC AR5, 2013) (Bowman, et al., 2013;Stevenson, et al., 2013).'

Page 3. Line 18. Text should clarify why the western Pacific is particularly opaque.

Page3, line 15 has been updated:

"For example, O3 changes in more opaque regions, e.g., the Western Pacific, a wet region due to convection, result in a much smaller change in TOA flux than in more transparent regions, e.g., the Middle East, a dry region due to downwelling (Kuai, et al., 2017)."

Page 6. Line 38. Reader wonders whether surface characteristics matter to OLR calculations, or are these captures by Ts?

Yes, surface characteristics like emissivity will effect the OLR calculations. For different surface types, like rock or vegetation, the emissivity could vary from 0.9 to 1. The surface type could relate to the differences of Ts but such difference would be much smaller than seasonal variation of Ts. Ts is a dominate factor to determine OLR. Here we are discussing the variability of the IRK for T_s is mainly controlled by T_s.

Page 9. Line 41. "The flux bias due to Ta is found to be negligible in all models, which indicates that the Ta is relatively accurate." The statement should probably be qualified to say simply that the modeled Ta estimates provide reasonable radiative fluxes at 9.6 μm. The temperatures may not be so accurate for other purposes.

We addressed the comments now at Page 10, line 5.
'The flux bias due to Ta is found to be negligible in all models, which indicates that the model Ta estimates provide reasonable radiative fluxes.'

Page 10. Lines 20-25. The authors might consider reordering the figures so that the figures showing biases in each driving variable come right after the figures showing the biases in the fluxes attributable to that variable – e.g., Figure 9 right after Figure 5. That way readers can more easily see how the driving variables affect fluxes.

We did reconsider the reviewer's great suggestion but decided to keep the current structure. Section '5.3 Vertically-resolved radiative bias of the O_3, H_2O and T' introduce the partitioned radiative biases for all four variables and summarized their features briefly. This let the readers to do the cross comparison between the variables. Then, in the section '5.4 The Spatial Source of TOA Flux Bias', we did the discussion in the style as reviewer's way. For example for O_3, we talk about Fig. 5 and Fig. 9. For H_2O, we discuss Fig. 6 and Fig. 10 together.

Page 11. Lines 20-25. Are these ozone biases, reported in ppb, weighted by air density? If not, perhaps they should be, given that the opacity of the atmosphere is affected by total ozone column, not average ozone mixing ratio in that column.

Yes, the ozone biases are reported in ppb. When we do the vertical integration in free troposphere (Eq. 5), the column density is considered.

Page 12. Line 8. What updates did Oman 2011 and Oman 2013 implement in GEOSCCM to improve the model's representation of ozone? Readers will want to know.

We added more detailed discussion about how does GEOSCCM improved their ozone representation in Page 12, line 26.

‘The GEOSCCM has been used to study the tropospheric O3 response to variations in the El Nino-Southern Oscillation (ENSO) where Oman, et al., (2011;2013) compared the model to satellite observations. These regular comparisons may have led to the improved simulation of tropospheric O3 profiles and consequently lower vertical O3 bias. The GEOSCCM model in the CCMI study uses the tropospheric/stratospheric chemical package developed within the Global Modeling Initiative (GMI) program (Duncan, et al., 2007) which has more realistic ozone chemistry, an internally generated quasi-biennial oscillation, an improved air/sea roughness parameterization and other improvements (Oman et al., 2014).

Nielsen et al., (2017) showed that GEOSCCM successfully reproduces the changes in the quasi-global (60°S–60°N) annual-mean trend in total O3 column since 1960s to the present day. For the present-day atmosphere, simulated tropospheric partial column O3 from GESCCM Ref-C1 for CCMI was compared to satellite observations of OMI and MLS (Ziemke et al., 2011). The differences are mostly a few Browner Dobson (DU) except the Northern Hemisphere subtropics and middle latitudes in autumn and winter with the 4–6 DU bias which are under investigation.’

New References:

Nielsen, J. E., S. Pawson, A. Molod, B. Auer, A. M da Silva, A. R. Douglass, B. Duncan, Q. Liang, M. Manyin, L. D. Oman, W. Putman, S. E. Strahan and K. Wargan, 2017. **Chemical mechanisms and their applications in the Goddard Earth Observing System (GEOS) earth system model**. Journal of Advances in Modeling Earth Systems, 9, 3019-3044, DOI: https://doi.org/10.1002/2017MS001011.

Molod, A., Takacs, L., Suarez, M., Bacmeister, J., Song, I.-S., & Eichmann, A. (2012). *The GEOS-5 Atmospheric General Circulation Model: Mean Climate and Development from MERRA to Fortuna* (NASA Tech. Rep. Series on Global Modeling and Data Assimilation, NASA/TM-2012–104606, Vol. 28, 117 pp.). Greenbelt, MD: NASA Goddard Space Flight Center.

Oman, L. D., & Douglass, A. R. (2014). Improvements in total column ozone in GEOSCCM and comparisons with a new ozone-depleting substances scenario. *Journal of Geophysical Research: Atmospheres*, 119, 5613– 5624. https://doi.org/10.1002/2014JD021590

Ziemke, J. R., Chandra, S., Labow, G. J., Bhartia, P. K., Froidevaux, L., & Witte, J. C. (2011). A global climatology of tropospheric and stratospheric ozone derived from Aura OMI and MLS measurements. *Atmospheric Chemistry and Physics*, 11, 9237– 9251. https://doi.org/10.5194/acp-11-9237-2011

Duncan, B. N., Strahan, S. E., Yoshida, Y., Steenrod, S. D., and Livesey, N.: Model study of the cross-tropopause transport of biomass burning pollution, Atmos. Chem. Phys., 7, 3713-3736, https://doi.org/10.5194/acp-7-3713-2007, 2007.

Page 12. Line 25. The text should provide a reference to substantiate the claim that MRI-EMS1r1 overestimates lightning NOx emissions and underestimates convective updrafts in the tropics.

We revised now Page 13, Line 4 as below
"Meanwhile the strong tropical upper tropospheric O3 biases in MRI-EMS1r1 are believed to be related to the weak tropical convective updraft and the large lightning NOx emissions in the model. The model with weak updraft fails to bring enough low O3 air from the surface to the upper troposphere in the tropics or overestimates the upper tropospheric mixing of stratospheric ozone-rich air. In addition, the global lightning NOx (LNOx) emission used in MRI-EMS1r1 is 10 TgN/yr. The best estimate of annual mean LNOx based on satellite data assimilation is 6.3 TgN/yr (Miyazaki et al, 2014). The LNOx in GEOSCCM is approximately 5 TgN/yr (Martini et al., 2011), which shows less tropical upper tropospheric O3 bias compared to MRI-EMS1r1. Thus, the overestimation of the O3 precursor in the upper troposphere is another reason for too much O3. Figure. A1 shows the improvement in the radiative biases due to less O3 bias in the experiment by half the LNOx emissions in MRI-EMS1r1 (see the appendix)."

Appendix

Here we compare the MRI-EMS1r1 experiment run: RefC1_50%LNOx with its RefC1 run. The new run's emission decreases by about 50% compared with the original run. The global lightning NOx emission annual mean in 2006 simulated in the experiment run is reduced from ~10.79 TgN/yr in RefC1 to ~5.21TgN/yr. The 10-year average changes from ~10.44 TgN/yr to ~5.18 TgN/yr.

We found the total flux bias is much reduced due to the improved O_3 radiative bias (Fig. A1 top two plots). The vertical resolved O_3 radiative bias shows as we expect, the strong overestimation of the tropical upper tropospheric O_3 radiative biases are much weaker in the new run (the middle two panels). This improvement is due to the less O_3 biases in this region due to reduced LNOx emission (the bottom plots).

We also see some changes in the latitudinal distributions of the H_2O radiative bias. This is because the reduction of the upper tropospheric O_3 will cause by the model responses in the O_3 heating rate, which would have radiative effect on the temperature, atmospheric stabilities, and convective activity (e.g. Newack et al. 2015). All these would impact water vapor and cloud.

New references:

Miyazaki, K., Eskes, H. J., Sudo, K., and Zhang, C.: Global lightning NO_x production estimated by an assimilation of multiple satellite data sets, Atmos. Chem. Phys., 14, 3277-3305, https://doi.org/10.5194/acp-14-3277-2014, 2014.

Martini, M., Allen, D. J., Pickering, K. E., Stenchikov, G. L., Richter, A., Hyer, E. J., & Loughner, C. P. (2011). The impact of North American anthropogenic emissions and lightning on long-range transport of trace gases and their export from the continent during summers 2002 and 2004. *Journal of Geophysical Research*, 116, D07305. https://doi.org/10.1029/2010JD014305

Nowack, P. J., Abraham, N. L., Maycock, A. C., Braesicke, P., Gregory, J. M., Joshi, M. M., Osprey, A., and Pyle, J. A.: A large ozone-circulation feedback and its implications for global warming assessments, Nat. Clim. Chang., 5, 41–45, doi:10.1038/nclimate2451, 2015. https://www.nature.com/articles/nclimate2451

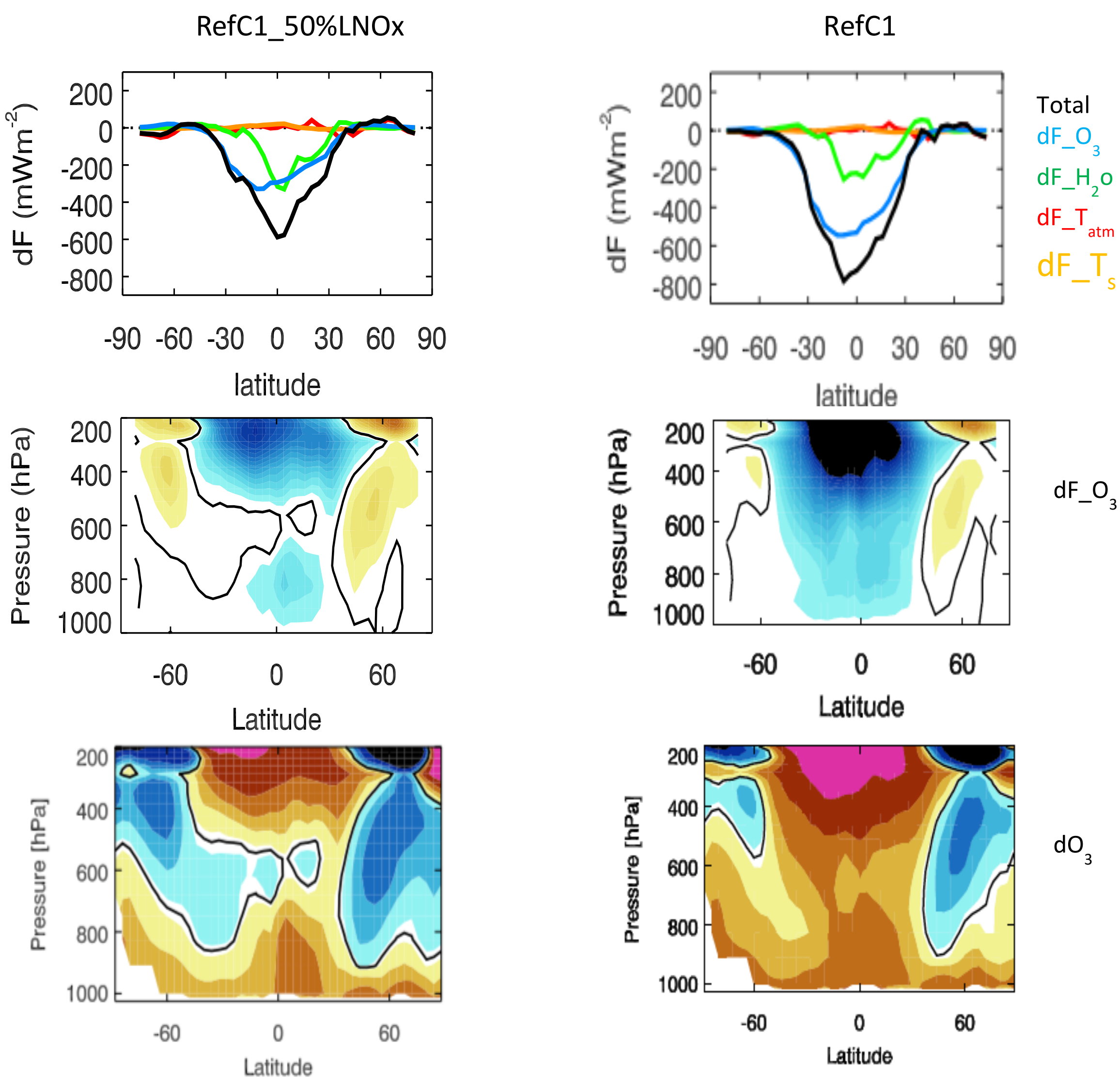

Fig. A1. The comparison of the MRI-ESM1r1 experiment of half LNOx in total flux bias (top row), O_3 radiative bias (middle row), and O_3 bias (bottom row). Left: new run with half LNOx. Right: RefC1 run. The black curves in the contour are the zero lines.

Page 12. Line 35. This reader does not understand "fact" 3, which gives this explanation for the bias in the radiative flux: "the systematic bias throughout the tropical troposphere, when propagate into the TOA flux, causes an accumulated bias in the radiative effect."

We revised the statement now at Page 13 Line 19 as below

'3) the simulations with the systematic bias throughout the tropical troposphere, when vertically integrated, accumulated into a larger column bias when compared to the models with vertically random biases.'

Page 13. Line 20. Why do the Ta biases in the tropics "shift between positive and negative vertically" in most models?

That means simulated air temperatures stay around the reanalysis profiles. These models better represent the air temperature than trace gases like H_2O and O_3.
We added the sentences in now Page 14, Line 7:

'The oscillated T_a biases suggest that simulated air temperatures stay around the reanalyzed profiles. These models better represent the air temperature than the trace gases like H_2O and O_3.'

Page 14. Lines 6-35. The text should clarify what of value is learned in the analysis of how biases in 9.6 μ band affect biases in the entire OLR band.

We added some statements for the value of the understanding about how biases in 9.6 μ band affect biases in the entire OLR band. Now Page 15, line 31:

'The anti-correlation between the biases in the 9.6-μm band and in the entire OLR band would suggest some bias drivers in the 9.6-μm band must play different roles at the other part of the OLR band. The further investigation of these processes would help to explain the radiative effect of different biases on the OLR estimations from models (Huang et al., 2008; Huang et al., 2014).
'

New References:
Huang, X. L., W. Yang, N. G. Loeb, *and* V. Ramaswamy *(*2008*),* Spectrally resolved fluxes derived from collocated AIRS and CERES measurements and their application in model evaluation: Clear sky over the tropical oceans, J. Geophys. Res., 113*, D09110, doi:*10.1029/2007JD009219.

Huang, X. L., Chen, X. H., Potter, G. L., Oreopoulos, L., Cole, J. N. S., Lee, D. M., & Loeb, N. G. *(*2014*)*. A global climatology of outgoing longwave spectral cloud radiative effect and associated effective cloud properties. Journal of Climate, 27*(*19*),* 7475–7492. https://doi.org/10.1175/JCLI-D-13-00663.1

Pages 14-16. The conclusion should emphasize that the paper focuses only on clear-sky OLR.

We added the statement for the clear sky in the following sentence. Now Page 15, line 31

'We have demonstrated a new method to quantitatively attribute the biases in O_3 band TOA flux from chemistry-climate model ensembles to O_3, H_2O, T_a, and T_s radiative components without cloud effect using observationally-constrained IRKs in the clear sky.'

Figure 4. The caption could say that the black curves are the same as the colored curves in Figure 3.

Added to the caption.

'Figure 4. The attribution of the total TOA flux bias for each model to four dominant components and their latitudinal distribution. The black curves are the same as the colored curves in Figure 3.'

Figure 5. What do the black curves represent?

We add the words as below

'Figure 5. Vertical resolved O_3 radiative bias. The black curves are the zero lines.

Figure 6. Vertical resolved H2O radiative bias. The black curves are the zero lines.

Figure 7. Vertical resolved T_a radiative bias. The black curves are the zero lines.

Figure 9. The zonal averaged vertical-latitudinal distribution of O_3 model biases to the TCR-1 O_3 assimilation data. The black curves are the zero lines.

Figure 10. The zonal averaged vertical-latitudinal distribution of H_2O biases (model to the ERA reanalysis data). The black curves are the zero lines.

Figure 11. The zonal averaged vertical-latitudinal distribution of T_a biases from models to the reanalysis data. The black curves in the contour are the zero lines.'

Figure 13. Equations should be shown only for those correlations and slopes that are statistically significant.

OK. We removed the equations of the fitting lines with weak correlations.

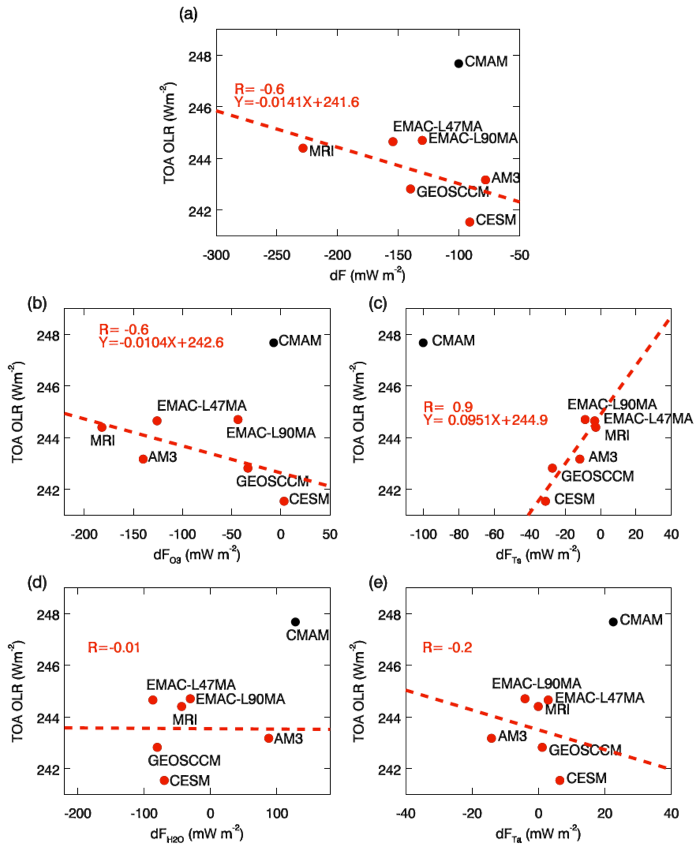

Figure 13. The correlation of the ozone band TOA flux biases to the model calculated broadband OLR (a) and the correlation of the attributed radiative components to the broadband OLR (b – e).

Table 3. Are the mixing ratios for ozone and water weighted by air density in each layer?

No. This is just the simple regional concentration average. We didn't compute the column bias by account for air density in each layer. The vertical weighting is considered while multiply with IRK and integral into tropospheric column.